

ASSESSMENT OF
ATMOSPHERIC CONDENSATION NUCLEI
ASSOCIATED WITH
JET AIRCRAFT TRAFFIC

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Annual Report on Contract No. N00014-76-C-0170, NR211-151

Submitted by

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ABSTRACT

Continued measurements of condensation nuclei (CN) in the size range $r \geq 0.01~\mu m$ by balloon-borne detectors have thus far allowed us to determine typical CN profiles as a function of latitude. We have also analyzed an observed high altitude jet particle layer to study the emissions and apply the results to other aircraft fleets. The larger particles ($r \geq 0.15~\mu m$) in the stratospheric sulfate layer have continued to decrease, although some transport effects have been apparent.

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INTRODUCTION

This report constitutes the annual report on ONR contract No. No0014-76-C-0170. Our main goal is to determine the source of atmospheric condensation nuclei (CN), the small particle portion of the atmospheric particle size distribution, and to assess the effects of jet aircraft on this particle distribution. In addition, regular measurements of the large sulfate aerosol particles have continued in an effort to assess the source and sink strengths of these particles which are important in the radiative energy balance of the atmosphere.

RESEARCH ACTIVITIES

Table 1 lists balloon soundings conducted since the last report (CN-7, April 1977). New results during the period (concerning CN) include a 300°C vaporization-flight and our first sounding from Brazil (5°S). The 300°C flight indicated that most of the tropospheric CN are not volatile at this temperature, suggesting a rather stable composition. The flight from Quixeramobim, Brazil, gave results similar to our previous equatorial soundings from Panama. The Brazil site is, however, much more workable and payload recovery did not present a problem.

Two reports are attached to this Progress Report which describe the CN observations in detail. They have both been submitted for publication.

In addition to the aforementioned research, we have continued monthly measurements of the $r \geq 0.15~\mu m$ stratospheric sulfate layer which has now decayed to a concentration of less than 1 cm⁻³ from a high of about 8 cm⁻³ in early 1975, shortly after the eruption of the volcano Fuego in Guatemala. This rather long period (~40 months) without any apparent large volcanic

eruptions has allowed us to study the decay of such injections which appear to have an exponential decay with a e^{-1} time of about 1 year for particles having $r \geq 0.15~\mu m$. Of particular interest here is if the concentration ever reaches a true "background" due to anthropogenic sources. Prior to the eruption of Fuego, the concentration had reached a low of about $0.5~cm^{-3}$ and has again reached this value recently. If no new volcanic injections occur during 1978, we should reach an all time record low aerosol concentration this summer. Small high altitude increases observed in late 1976, early 1977, are believed to be due to transport of aerosol from equatorial regions during this period. Data from Brazil is consistent with this view.

TABLE 1

Flight No.	Location	Date	Constituents Measured
W-143	Daramie	17 March 1977	Α, Τ
P-12	Panama	27 March 1977	CN, T
P-13	Panama	31 March 1977	CN, T
W-144	Laramie	6 April 1977	A(2), T
W-145	Laramie	28 April 1977	А, Т
W-147	Laramie	6 May 1977	A, CN, T
W-148	Laramie	12 May 1977	Α, Τ
W-149	Laramie	1 June 1977	IC, T
W-150	Laramie	14 June 1977	Α, Τ
W-151	Laramie	6 July 1977	А, Т
W-152	Laramie	28 July 1977	Α, Τ
W-154	Laramie	12 August 1977	А, Т
W-155	Laramie	19 August 1977	ID, T
W-156	Laramie	26 August 1977	CN, T
W-157	Laramie	1 September 1977	Λ, Τ
W-159	Laramie	13 September 1977	CN, T
W-160	Laramie	29 September 1977	A, SC, T
W-161	Laramie	7 October 1977	Α, Τ
W-162	Laramie	12 October 1977	ID, T
W-163	Laramie	12 October 1977	IC, SC, T
W-164	Laramie	20 October 1977	ID, T
W-165	Laramie	20 October 1977	IS, SC, T
W-166	Laramie	3 November 1977	Α, Τ
W-167	Laramie	14 December 1977	O(3), T
W-168	Laramie	15 December 1977	Λ, Τ
A-30	McMurdo, Antarctica	17 January 1978	A, T
W-169	Laramie	22 January 1978	A, T
A-31	South Pole	23 January 1978	CN, T
W-170	Laramie	24 February 1978	ID, T
W-171	Laramie	1 March 1978	IC, SC, T

Continued

TABLE 1

Flight No.	Location	Date	Constituents Measured
W-172	Laramie	8 March 1978	А, Т
Q-3	Quixeramobim, Brazil	8 March 1978	A, T
Q-4	Quixeramobim, Brazil	9 March 1978	CN, T
W-173	Laramie	28 March 1978	A, SC, T
W-174	Laramie	20 April 1978	A, T

A - Aerosol $(r \ge 0.15 \mu m)$

CN - Condensation Nuclei (r \leq 0.01 $\mu\text{m})$

T - Temperature

ID - Ion Density

IC - Ion Chamber

SC - Scintillation Counter

0 - Ozone

BALLOON OBSERVATIONS OF A PARTICLE LAYER INJECTED BY A STRATOSPHERIC AIRCRAFT AT 23 KM

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Abstract. An extensive layer of small particles ($r \gtrsim 0.01~\mu m$) was observed in association with the passage of a jet aircraft at 23 km altitude. The data are used to estimate the small particle emission index and the source strength of such aircraft at stratospheric altitudes. The impact of such flights on the stratospheric particle population is probably negligible; however, when the results are applied to the currently projected Concorde SST fleet in the stratosphere and the commercial jet fleet in the upper troposphere, both are estimated to contribute non-negligible equilibrium concentrations of such particles to their respective altitude regimes.

Observations

Balloon soundings of very small particles, known as condensation nuclei (CN), have been conducted by our group since 1973. Purposes of the measurements are to determine the CN source, global, distribution and their importance as seed particles for growing the larger particles of the stratospheric aerosol layer.

The CN detector consists of a thermal gradient growth chamber and an optical individual particle detector. The detector system is sensitive to particles with radii $\gtrsim 0.01$ µm, weighs about 8 kg and is flown to an altitude of 27 km by a 3 kg rubber sounding balloon. For further information on the detector system, its characteristics and typical concentration profiles, see Rosen and Hofmann (1977).

To date, 27 CN soundings have been conducted mainly from Laramie, Wyoming (41°N) but also from Fairbanks, Alaska (65°N), Panama (9°N), Quixeramobim, Brazil (5°S), Mildura, Australia (34°S), McMurdo Station, Antarctica (77°S) and the South Pole. It is important to point out that in all these flights, while tropospheric CN concentrations were high and somewhat variable (200 cm⁻³- 1000 cm⁻³), stratospheric levels were low and very uniform globally with concentrations of about 20 cm⁻³ at 15 km, decreasing smoothly to about 2 cm⁻³ at 25 km. (Rosen and Hofmann, 1977; Rosen et al, 1978).

On some of the Laramie soundings, two CN detectors were flown, one in a normal configuration and one with the air intake tube heated to a constant temperature to determine the degree of volatility of the CN at that temperature. On one of these occasions (September 18, 1976) an unusual 0.2 km thick layer of CN was detected at an altitude of 23 km on both balloon ascent and parachute descent by both detector systems. The concentration of particles with radii $\geq 0.01~\mu m$ in the layer reached 85 cm⁻³ above a normal background of about 2 cm⁻³ at this altitude. The vertical CN profiles, as obtained by the two detectors on ascent, are shown in Figure 1. The temperature chosen for the heated intake detector on this sounding was 150°C, hot enough to boil an aerosol consisting of a 75% sulfuric acid - 25% water mixture, a likely stratospheric aerosol composition (Rosen, 1971; Toon and Pollack, 1976), but not hot enough to dissociate compounds such as ammonium sulfate.

The layer exhibited considerable structure as can be seen in Figure 2, where data for the 22.5 - 22.9 km region are shown with higher resolution (points below and above a concentration of 50 cm⁻³ were calculated from a registration of 512 and 1024 particles, respectively; thus statistical fluctuations are on the order of 4.5 and 3%, respectively). Since data for the heated intake detector were commutated with other information, portions of the heated CN record are missing (e.g., the region around 22.7 km in Figure 2). However, it appears that to a major extent, the CN in the layer was volatile at 150°C. Figure 1 shows that except for certain tropospheric layers (which may partially contain water-bearing aerosol), condensation nuclei in the atmosphere are generally not volatile at 150°C.

Since we had never observed a CN layer in the stratosphere in any of our other soundings, and since the observation was confirmed by two detectors on ascent and descent, the observation is believed to be significant. The layer was probably caused by some transient phenomenon such as a jet aircraft. A large meteor could conceivably leave a detectable trail of vaporized material down to 20 km, but this possibility appears most unlikely in view of the volatility of the particles in the layer.

Through the Office of Naval Research and the Federal Aviation Administration, it was determined that on 17 September 1976, a high altitude military aircraft flew a training mission in the vicinity of Laramie. The aircraft was traveling at a speed of about mach 2.5 (1650 knots) and had a point of closest approach to Laramie about 150 km to the northwest with an altimeter reading of 76,000 ft (~23 km) at about 1840 Z, 17 September 1976. The time of penetration of the layer on balloon ascent was about 1300 Z, 18 September 1976, or about 18 hours after the aircraft had passed.

Meteorological conditions at this altitude for this period were dominated by a high pressure system centered over the northwest corner of the United States. Winds in the region of Laramie, and the aircraft path at 23 km were light (~5 knots) and variable (generally out of the north) so it is most probable that we encountered the remnants of a high altitude jet exhaust trail. This constitutes, to the best of our knowledge, the first in situ detection of the passage of a stratospheric aircraft flying above 20 km. Beyond this, the observation can be used to estimate some of the diffusive properties of the stratosphere and to study the characteristics of such stratospheric emissions.

Discussion

In terms of the vertical eddy diffusion coefficient in effect at the time, we can estimate a lower limit by assuming the measured 0.2 km thickness was typical of the entire plume. For radial diffusion from an instantaneous cylindrical source, the data indicate a diffusion coefficient of 400 cm²sec⁻¹. Since typical average accepted values at this altitude are about 5000 cm²sec⁻¹ (Johnston et al, 1976) one might conclude that we were sampling only the edge of the plume. However, it is also possible that the low value measured here is indicative of the somewhat stagnant meteorological conditions in effect at the time and suggests that such direct measurements of diffusion parameters need to be made under diverse conditions, seasons and at all altitudes in order to be a valid method of obtaining an average profile.

From available data, we can make an order of magnitude estimate of the number of particles having $r \gtrsim 0.01~\mu m$ emitted per gm of fuel expended. We can also estimate the source strength of such aircraft flights, i.e., the contribution to the natural background of these particles.

It appears that the only US aircraft capable of the performance indicated by the information obtained from the 17 September training mission is the SR-71 (mach 3 strategic reconnaissance aircraft). This aircraft's twin engines consume about 30,000 l/hr of JP-7 fuel when cruising near mach 3 at 24 km (Jane's All the World's Aircraft, 1975).

We can estimate a lower limit cross section of the exhaust plume from the measured vertical thickness and from horizontal diffusion parameters. The latter are not well known and are a major source of uncertainty in this analysis. From the data compiled by Bauer (1974), we estimate that at 23 km, in the 18 hour injection to detection time, the jet effluent plume would have spread horizontally a distance of from 20 - 90 km.

Taking an observed average particle concentration of 50 cm $^{-3}$ in a 0.2 km thick layer, we calculate that $(2\text{-}9)\text{x}10^{13}$ particles having $r \geq 0.01$ µm were emitted for each gram of fuel expended. Most of these particles probably have radii < 0.1 µm. If we use an average radius of 0.03 µm and a particle specific gravity of 1 gm cm $^{-3}$, we find an emission index (g aerosol per kg fuel) of 2-10, i.e., 0.2 - 1% of the mass of the fuel expended appears as particles. Because of the necessary assumption of a particle size, the latter number is very uncertain. Typical jet engines operating at ground level have emission indexes of from 0.1 - 3 (Sawyer, 1970), while the exhaust trail of an F-104 jet aircraft at an altitude of about 9 km indicated an emission index of 1.5 (Rosen and Greegor, 1974). Because of the uncertainties involved, it is difficult to say if the emission index is higher in the stratosphere. It does, however, appear to be no lower than at lower altitude. Different engines and different fuels also complicate the issue. Further inaccuracies in the

emission index estimate are introduced due to the fact that we cannot detect particles having radii less than 0.01 µm. Although such particles may not contribute significantly to the total mass, they may be important in forming larger particles through coagulation and in gas-particle interactions in the stratosphere.

To estimate a particle source function, knowledge of the flight frequency is necessary. This is probably impossible to determine but from the frequency of sonic booms in the Laramie area, we estimate a minimum frequency of one flight per week. If the flight we encountered was typical (about 4 x 10 kg of fuel was expended), then $(4-20)x10^{22}$ particles having $r \geq 0.01$ µm are deposited each year. Spreading these out uniformly over the northern hemisphere in a stratospheric layer 10 km thick, we find a source strength of 0.02 - 0.07 particles cm⁻³ yr⁻¹. Since the stratospheric lifetime, due mainly to mixing, is of the order of one year, such aircraft contribute about 0.02 - 0.07 particles cm⁻³ to the stratospheric background concentration. Current levels of $r \geq 0.01$ µm particles at 23 km are in the range of 2-5 cm⁻³ (Rosen and Hofmann, 1977), nearly two orders of magnitude greater than our estimate for stratospheric flights with a frequency of one per week. Even if the plume was actually several times thicker than measured, the effect on the stratosphere would be negligible.

It is of interest to apply the results to the projected Concorde SST fleet (16 aircraft). The Concorde at mach 2 cruise burns fuel at about half the rate (16,800 kg/hr) as the SR-71 and does so at lower altitudes (15-18 km) where the background CN concentration is about 5 times higher (10-25 cm $^{-3}$) than at 23 km (Rosen and Hofmann, 1977). Broderick et al (1975), in the final

Climatic Impact Assessment Program (CIAP) report, estimate that the average useage of each supersonic aircraft of the Concorde type will be 4.5 hours per day above 15 km altitude by 1980 or before. This would result in about 4.4 x 10⁸ kg/yr of fuel expended between 15 and 18 km. This may be compared with the SR-71 estimate of 2 x 10⁶ kg/yr for weekly flights, and we see that if the Concorde engines emitted particles as the SR-71 engines, then 4-16 particles cm⁻³ could result at equilibrium, i.e., of the same order of magnitude as the current background of 10-25 cm⁻³ in this altitude region. However, an altitude of 15 km is low enough so that loss effects due to the presence of the tropopause, especially in summer (the tropopause height varies with season between about 9 and 15 km at mid-latitudes), might shorten the effective lifetime of the particles.

We can look further to standard commercial jet aircraft operating predominantly in the 35,000 - 39,000 ft (10.7 - 11.9 km) range where the background CN concentration varies from about 200 - 1000 cm⁻³ (Rosen and Hofmann, 1977). Broderick et al (1975) estimate that about 3 x 10¹⁰ kg/yr of fuel were consumed at altitude by the world fleet (commercial and military) in 1970. Using this figure and reducing the average lifetime by a factor of ten (to 0.1 yr) for the largely tropospheric particles, we arrive at an equilibrium particle concentration of 25 - 105 cm⁻³. We must remember that all these ranges of expected concentrations are in effect lower limits since we may have been sampling only the edge of the plume. It appears that the present tropospheric aircraft fleet may be contributing to the natural background of small particles in the upper troposphere, but the degree is probably not sufficient to be easily discerned with the few measurements available.

The earliest measurements of CN in the upper troposphere were made by Junge et al (1961) in 1959, in the early years of the jet age. Only a few balloon soundings were conducted indicating levels of about 200 particles cm⁻³; however, it is impossible to arrive at a reliable average background profile for that period with the available data. There were no measurements of CN in the upper troposphere between 1959 and 1973. An increase in the average background level of 100 particles cm⁻³ during this time interval cannot be ruled out by the data. The suggestion of increased tropospheric concentrations since 1959 and the extensive fine structure in the tropospheric CN profile led Rosen et al (1974) to suggest a jet air-traffic source for these particles as early as 1974.

Other evidence suggests that if there is a tropospheric air traffic CN contribution, it probably amounts to no more than 100 cm⁻³. Soundings made in equatorial regions and in the southern hemisphere (Rosen and Hofmann, 1978) suggest that the tropospheric concentration of CN is not drastically different in the southern hemisphere. One concludes that tropospheric jet aircraft (flights of which are more numerous in the northern hemisphere) are not a dominant source of tropospheric CN. Again, average differences between hemispheres of 100 cm⁻³ cannot be ruled out with the data at hand.

The observation of the volatility of the aerosol particles is important since it is related to particle composition. The fuel used, JP-7 in this case, is a special low vapor pressure hydrocarbon with a sulfur content similar to ordinary jet fuel, i.e., a maximum percentage by weight of 0.1 (emission index of 1) where the emitted sulfur compound is mostly SO_2 . In view of the observed volatility of the produced aerosol, and sufficient availability of SO_2 in the jet exhaust, it is highly likely that the aerosol is at least

partially composed of sulfuric acid. Larger particles ($r \ge 0.1~\mu\text{m}$), occurring naturally in the stratospheric aerosol layer, have a similar composition thought to be due to gas to particle conversion with volcanic $S0_2$ probably constituting the major gaseous source. If the particles which form in the jet exhaust trail are indeed sulfuric acid droplets, then their production could be reduced by reducing the sulfur content of the fuel.

What impact, if any, might an increase in small particles have? It is well known that larger particles, in the optically active size range (r \geq 0.1 μ m), can affect incident solar radiation if present in the stratosphere in sufficient concentrations. Most current models of how these particles form in the stratosphere (Turco et al., 1976; Rosen et al., 1977) involve gas to particle conversion by condensation on existing nuclei with the gas($\rm H_2SO_6$ vapor derived from $\rm SO_2$) provided by volcanoes or anthropogenic sources and the condensation nuclei provided by diffusion from the troposphere where they are found in large numbers. Thus, CN probably play an important role in the formation of optically active aerosol in the stratosphere. Since the stratospheric concentrations of CN have been observed to be rather low and remarkably constant, (Rosen and Hofmann, 1977), it will be important to continue to monitor them as the stratosphere and upper troposphere see increased usage as a transportation medium.

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References

- Bauer, E., Dispersion of tracers in the atmosphere and ocean; survey and comparison of experimental data, J. Geophys. Res. 79, 789, 1974.
- Broderick, A. J., C. S. Downie and R. C. Oliver, Aircraft engine emissions at altitude, Propulsion Effluents in the Stratosphere, CIAP Monograph 2, Ch. 7, U.S. Department of Transportation publ. DOT-TST-75-52, September, 1975.
- Jane's All the World's Aircraft, 1974-75, Jane's Yearbooks, London, 1975.
- Johnston, H. S., D. Kattenhorn and G. Whitten, Uses of excess carbon 14 data to calibrate models of stratospheric ozone depletion by supersonic transports, J. Geophys. Res. 81, 368, 1976.
- Junge, C. E., C. W. Chagnon and J. E. Manson, Stratospheric aerosols, J. Meteorol. 18, 81, 1961.
- Rosen, J. M., The boiling point of stratospheric acrosols, <u>J. App. Meteorol. 10</u>, 1044, 1971.
- Rosen, J. M. and R. Greegor, Jet engine soot emission measured at altitude, J. of Aircraft 11, 243, 1974.
- Rosen, J. M., R. G. Pinnick and R. Hall, Recent measurements of condensation nuclei in the stratosphere, Proc. Third Conf. Climatic Impact Assessment Program, U.S. Department of Transportation publ. DOT-TSC-OST-74-15, p.298, November, 1974.
- Rosen, J. M. and D. J. Hofmann, Balloonborne measurements of condensation nuclei, J. App. Meteorol. 16, 56, 1977.
- Rosen, J. M., D. J. Hofmann and P. Singh, A stratospheric aerosol model with perturbations induced by the space shuttle particulate effluents, Univ of Wyoming Atmospheric Physics Report AP39, July, 1977.
- Rosen, J. M., D. J. Hofmann and K. H. Kaselau, Vertical profiles of condensation nuclei, Univ of Wyoming Atmospheric Physics Report AP-41, January, 1978.
- Sawyer, R. F., Reducing jet pollution before it becomes serious, J. of Astronautics and Aeronautics 8, 62, 1970.
- Toon, O. B. and J. B. Pollack, A global average model of atmospheric aerosols for radiative transfer calculations, J. App. Meteorol. 15, 223, 1976.
- Turco, R. P., P. Hamill, O. B. Toon and R. C. Whitten, A model of stratospheric aerosols, Atmospheric Aerosols: Their Optical Properties and Effects, available through NTIS, NASA CP-2004, 1976.

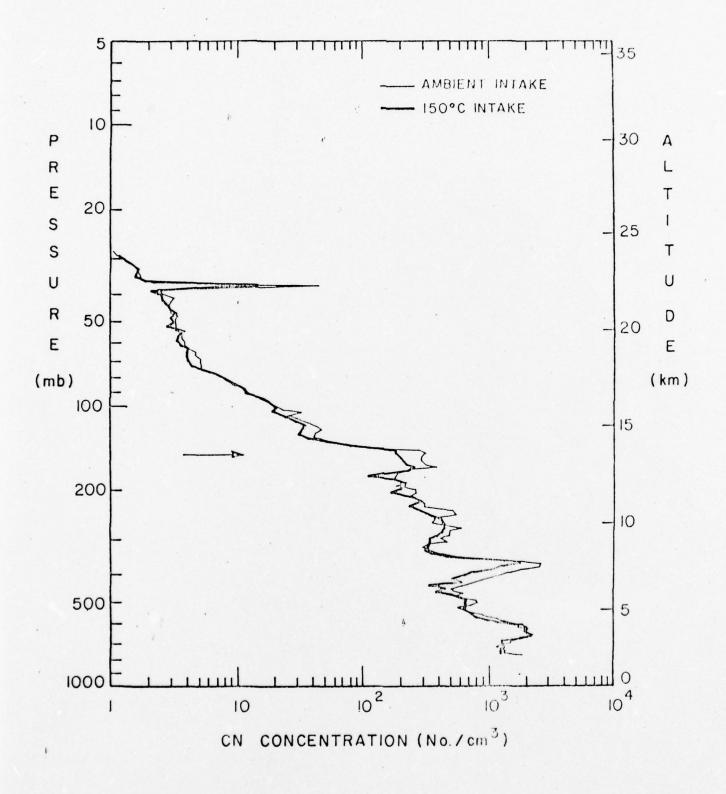


Figure 1. Condensation nuclei (CN) concentration versus height at Laramie on 18 September, 1976. The arrow marks the observed position of the tropopause.

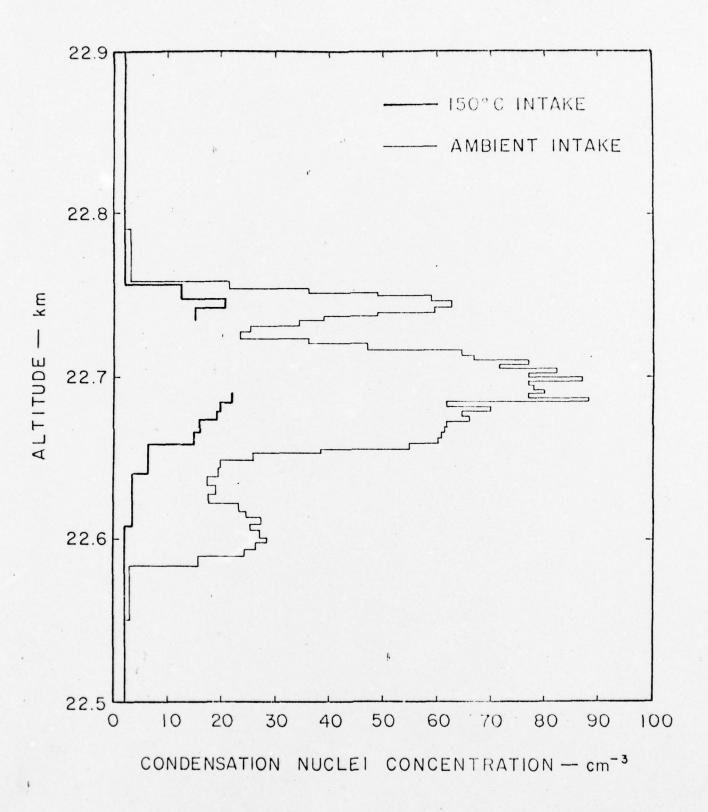


Figure 2. The 22.5-22.9 km region of figure 1 in greater detail.

VERTICAL PROFILES

OF

CONDENSATION NUCLEI

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Introduction

In an earlier paper (Rosen & Hofmann, 1977) we presented the results of a number of condensation nuclei (CN) soundings made over Laramie using an experimental thermal gradient diffusion cloud chamber (TGDCC). The purpose of this note is to describe further developments in the evaluation of the TGDCC CN counter and present the vertical profiles as measured with the instrument at several locations in both the northern and southern hemisphere.

Progress in CN Counter Development and Evaluation

In the early evaluation of the TGDCC no conclusive evidence could be established that above 20 km the supersaturation (~ 10%) was high enough to produce growth on all the particles and hence it was not known whether all of the CN present were being counted. To further investigate this aspect of the counter, simultaneous comparison soundings were made with an expansion type CN counter developed by one of the authors (Käselau, 1974) that was operated at a much higher supersaturation (~ 200%) than the TGDCC. It should be mentioned that the working fluid in the expansion type CN counter was water while in the other counter it was glycol. Three soundings were made with both instruments on the same balloon. The results of the average of all three soundings are shown in figure 1. Most of the data has been smoothed somewhat by taking a seven point running average.

Below 15 km it is clear that there is good relative agreement between the two instruments. The slight discrepancy in the absolute values may be attributed to the diversity of the two counters and the general problem of obtaining accurate absolute values. Above 15 km there is lack of agreement in the fine structure but a general agreement in the absolute values within about a factor of two is apparent. The reason for lack of agreement in the fine structure above 15 km can at least partially be attributed to the large

expected statistical flucuations in the expansion chamber measurements; at high altitude very few and frequently no particles are observed after the expansion. This would obviously produce large excursions in the apparent vertical profile.

In the earlier efforts to use the TGDCC at high altitude it was found that the instrument ceased to function altogether above about 25 km. It was later found by decreasing the supersaturation, the instrument continued to function to a much higher altitude. The TGDCC data shown in figure 1 above 23 km was obtained on one of the three comparison flights with a reduced supersaturation. These results have led us to the tentative conclusion that the relatively low supersaturations employed in the TGDCC are still high enough to allow the instrument to count at least a large fraction of the CN present even to as high an altitude as 30 km.

CN Profiles at Other Latitudes.

In addition to the measurements made in Laramie a number of soundings have been made at other locations. Table 1 contains a summary of this activity. Before a meaningful examination of the latitudinal variations can be made, typical profiles at each site must be identified by comparing the results of several soundings. From table 1 it is clear that a very limited number of soundings at the field sites have been made and hence there may be some question as to whether the profiles presented here are representative. However, in view of the fact that there seems to be a good deal of consistency in the present data and that additional soundings at each site may not be forthcoming, we feel that it is justifiable at this time to present and discuss the results so far obtained.

Evidence that the present set of profiles is at least somewhat typical can be found in figures 2 through 5. Figure 2 shows a comparison of three soundings made over Panama and within certain range of variation they all appear quite similar. A comparison of two soundings made over Fairbanks, Alaska is shown in figure 3 and except for the layer at 10 km (to be discussed later) they are also quite similar. Even soundings made in the arctic (Fairbanks) and antarctic region

(McMurdo), as compared in figure 4, show a great deal of similarity. However, the two antarctic soundings illustrated in figure 5 show a large difference in mixing ratio in the troposphere. At the present time (for reasons discussed below) we believe that the high values of mixing ratio observed in the south pole profile in Figure 5 is non-typical and probably related to advection of mid-latitude air with its associated higher CN content. The 10 km layer in one of the Fairbanks soundings (figure 3) and also the 4 km layer in the McMurdo sounding (figure 5) could lend themselves to this same interpretation.

The single sounding made over Mildura, Australia is shown in figure 6. Although it is not strictly possible to determine whether or not this is a typical sounding, it does appear quite similar to the equatorial soundings made over Panama.

Discussion and Conclusions

The relatively high CN mixing ratios in the troposphere are indicative of a tropospheric source. A decrease in CN concentration is generally observed near the local tropopause and frequently it is rather dramatic. The equatorial and mid-latitude troposphere apparently exhibit a CN mixing ratio about an order of magnitude larger than polar regions. This could mean that high latitude regions act as an effective net sink for CN. Another explanation of the relatively clean arctic troposphere may be related to possible subsidence of stratospheric air through the high latitude tropopause, a process that would tend to keep the high arctic flushed with air of relatively low CN mixing ratio. However, it is not clear if there would be enough available mass in the stratosphere to keep the the arctic region purged with clean air. The mass of the northern hemisphere stratosphere is about four times the tropospheric mass above 65°N. If the half life of stratospheric air is approximately I year and half of the turnover is available for subsidence through the arctic tropopause, then only about one clean air change per year in the arctic troposphere could be expected. This may not be sufficient to maintain the observed low CN concentration.

In tropospheric regions where the polar and lower latitude air masses mix, large fluctuations may be expected; polar air would be characterized by a low CN mixing ratio while air masses from mid-latitude would display a relatively high mixing ratio. As previously mentioned, the high latitude CN soundings so far obtained do in fact suggest the presence of two distinct air masses each characterized by their CN mixing ratio. For instance, the layer at 10 km in figure 3 is characterized by a CN mixing ratio typical of mid latitude air which has led us to tentatively identify it with a lower latitude origin. Other researchers have also seen these same types of fluctuations in the polar CN concentration (Hogan, 1975; Flyger et al, 1973). Thus CN may prove to be a valuable tracer of tropospheric air masses in polar regions.

The large difference in the characteristic CN mixing ratio between the stratosphere and troposphere suggests that here too CN may prove to be a useful tracer. A tropospheric air mass transported into the stratosphere would exhibit a much larger CN mixing ratio than the surrounding air and hence it could easily be identified until extensive mixing destroyed its original character.

Figure 7 is a composite of soundings made at 5 different latitudes. The variation of the profiles with latitude, as governed by the tropopause height, can easily be recognized. It is also clear from this figure that above 20 km all profiles display approximately the same constant mixing ratio. If the mixing ratio remains constant to higher altitudes, as the figure suggests, then an extraterrestrial or very high altitude source for the stratospheric CN would be probable. Some researchers, however, (Junge et al, 1961) have considered a tropospheric source for CN and successfully described the lower stratospheric profile by applying coagulation - diffusion processes. In the high altitude limit of this argument, the CN mixing ratio should decrease because sedimentation becomes large enough to prevent the particles from being well mixed.

Thus, extending the measurements to higher altitudes will help to determine the source of stratospheric CN. Studies of the chemical composition and size may also be of considerable help but they are much more difficult to carry out for the very small particles under consideration here.

In conclusion, even though the TGDCC CN counter is still in the development stage, we believe it shows enough promise, in terms of producing consistant and interpretable profiles, to warrant its further use and development. In addition, the measurement of CN appears to offer great and untapped potential in the field of atmospheric tracers.

Acknowledgments.

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TABLE
Summary of Condensation Nuclei Soundings

Number of Soundings	Date of Soundings	Tropopause Height (km)
1	1/19/77	10.5
1	1/19/76	8.3
1	2/3/77	15.0
3	4/1/76	16.0
*	3/27/77	17.9
	3/31/77	16.8
2	5/14/76	8.0
	5/16/77	9.3
17	12/19/73-9/13/77	9-15
	Soundings 1 1 1 3	Soundings Soundings 1 1/19/77 1 1/19/76 1 2/3/77 3 4/1/76 3/27/77 3/31/77 2 5/14/76 5/16/77

References

- Flyger, H. K., Hansen, W. J., Megaw and Cox, L. C., 1973: The Background Level of the Summer Tropospheric Aerosol over Greenland and the North Atlantic Ocean, J. Appl. Meteor., 12, 161-174.
- Hogan, Austin W., Antarctic Aerosols. 1975: J. Appl. Meteor., 14, 550-559.
- Junge, C. E., Chagnon, C. V. and Manson, J. E., 1961: Stratospheric Aerosols, J. of Meteor. 18, 81-108.
- Käselau, K. H., 1974: Measurements of Aerosol Concentration up to a height of 27 km., Pure Appl. Geophys., 112, 877-885.
- Rosen, J. M. and Hofmann, D. J., 1977: Balloon Borne Measurements of Condensation Nuclei, J. Appl. Meteor. 16, 56-52.

Figure Captions

- Figure 1. A comparison of the average of 3 profiles as simultaneously measured by two different types of CN counters. The flights were made on September 18, 22 and 25, 1976, over Laramie. The thin line refers to the expansion chamber type counter while the thick line refers to the thermal gradient optical counter. The mixing ratio is in units of number of CN per milligram of air.
- Figure 2. A comparison of 3 soundings made over Panama. Reading from left to right at the 175 mb level, they were made on the following dates: 4/1/76; 3/27/77; 3/31/77.
- Figure 3. A comparison of 2 soundings made over Fairbanks, Alaska. Reading from left to right at the 300 mb level they were made on 5/14/76 and 5/16/76.
- Figure 4. A comparison of a sounding made at Fairbanks, Alaska (5/14/76) and one made at McMurdo, Antarctica (1/19/76). The McMurdo sounding shows a narrow layer at 600 mb.
- Figure 5. A comparison of the McMurdo, Antarctica sounding and the South Pole sounding. The South Pole sounding has higher values in the troposphere.
- Figure 6. The profile of CN obtained over Mildura, Australia on Feb. 3, 1977.
- Figure 7. A comparison of what is believed to be typical soundings from 5 different latitudes. Reading from left to right at the 90 mb level they are: McMurdo, Antarctica 1/19/76; Fairbanks, Alaska 5/14/76; Laramie, Wyoming 9/25/76; Mildura, Australia 2/3/77 and Panama, 3/31/77.

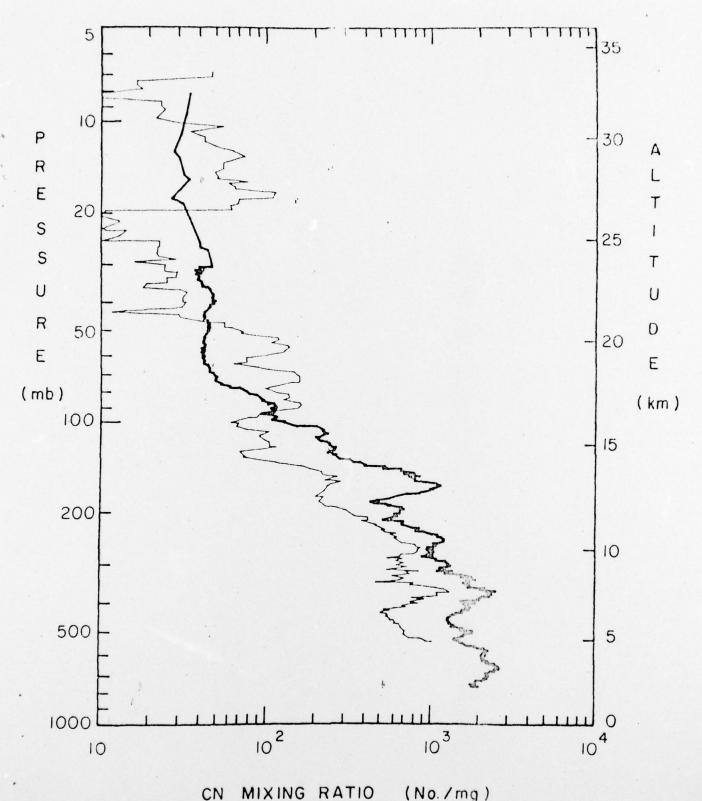
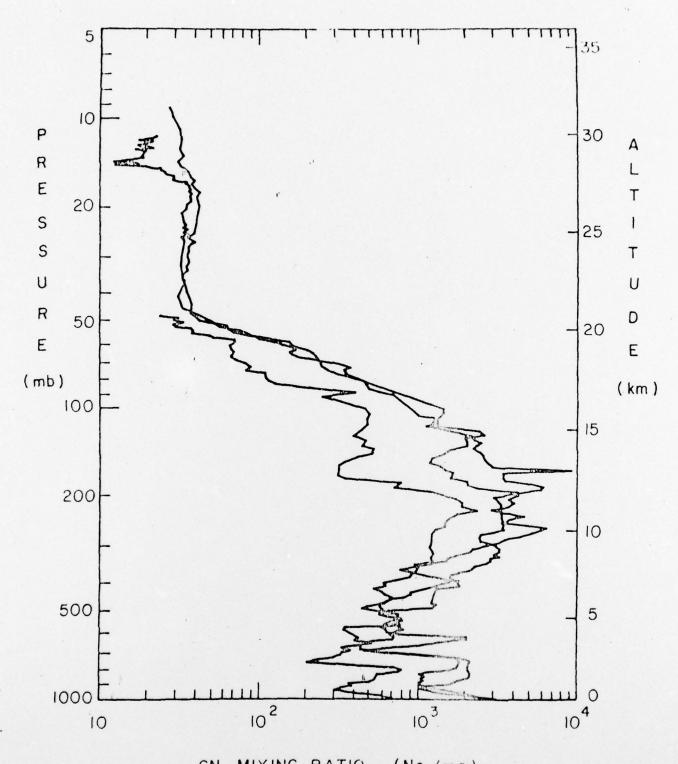
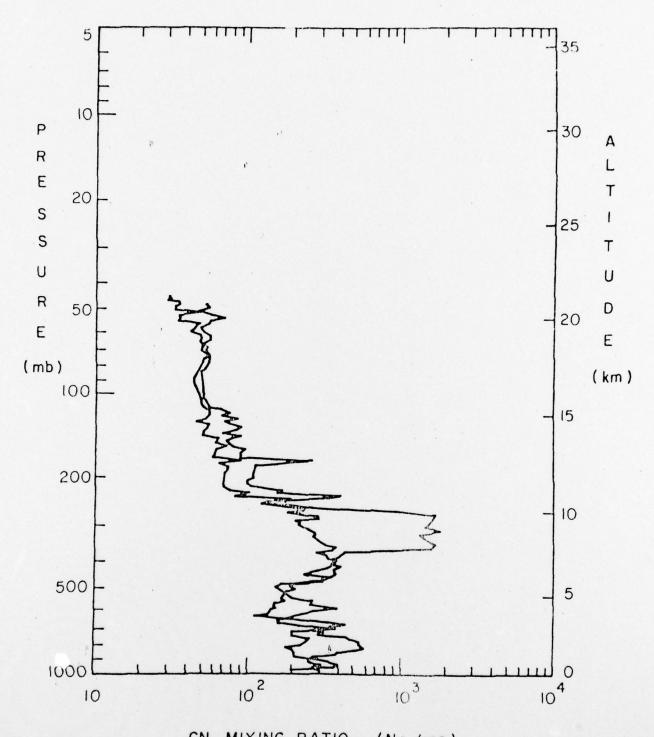


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CN MIXING RATIO (No./mg)
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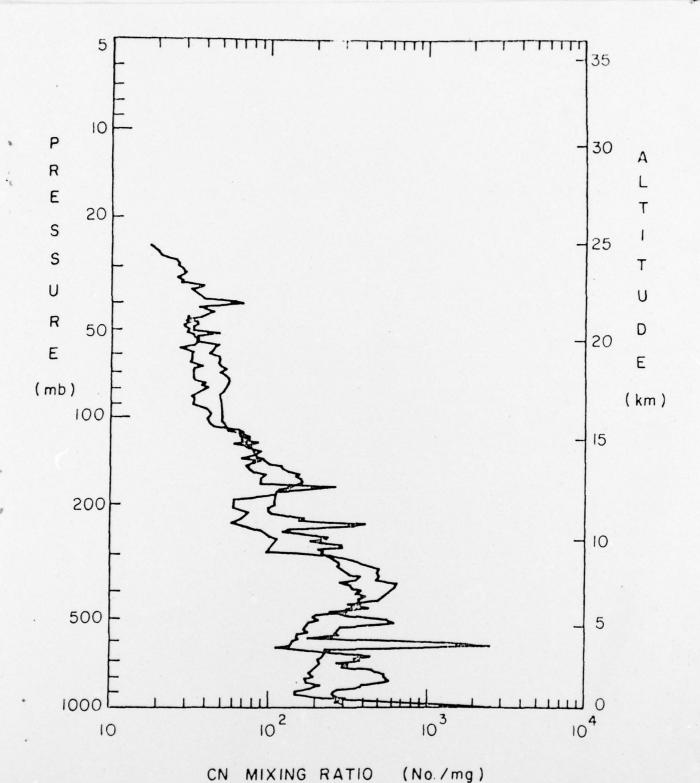
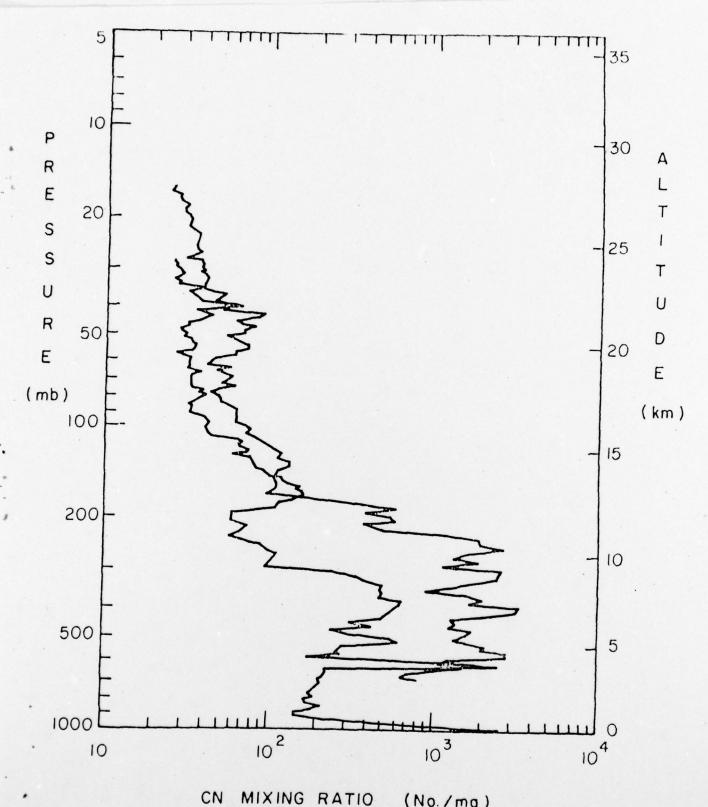


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The South Pole sounding has higher values in the troposphere.

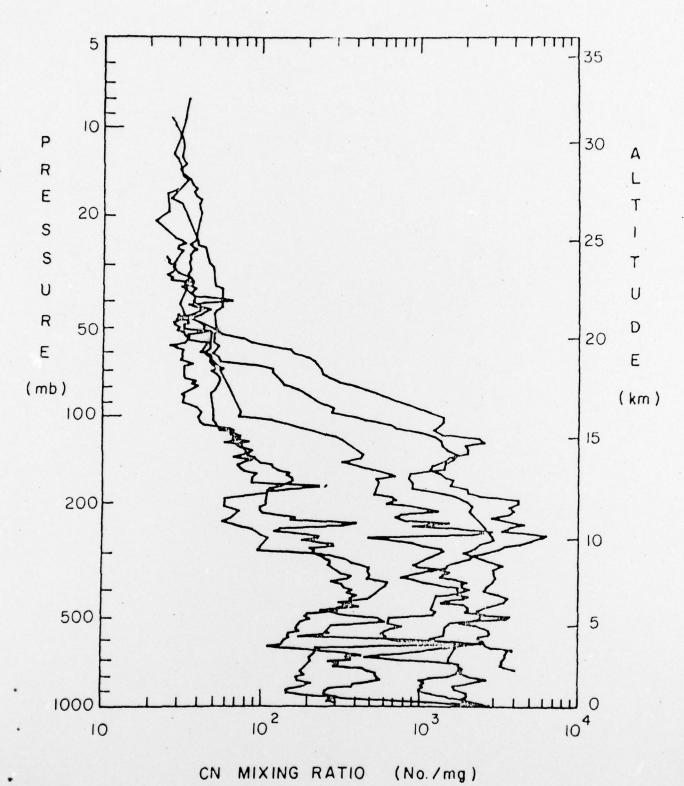


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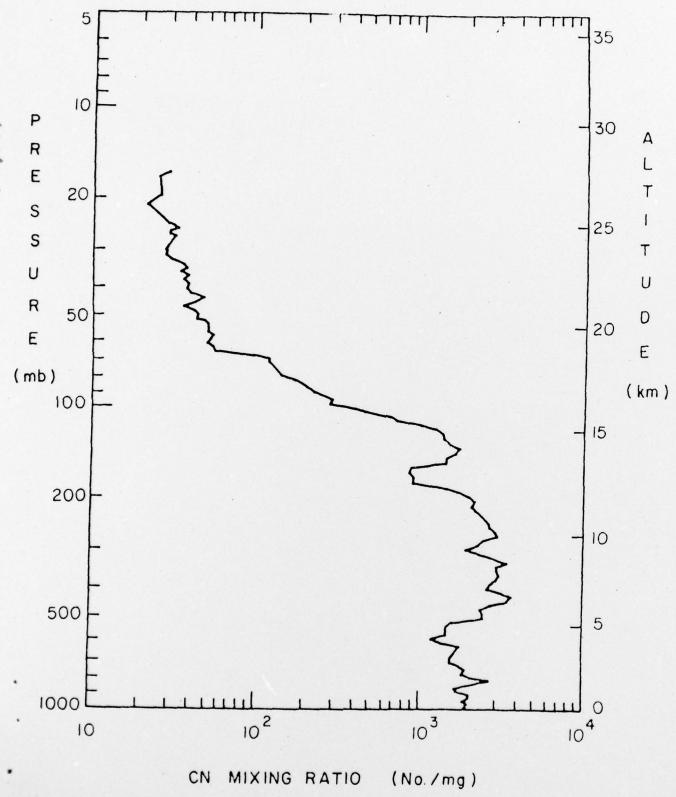


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